ON THE THEORY OF THE COHERENT SPONTANEOUS EMISSION

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Some problems of the theory of coherent spontaneous radiation are considered. It is shown that the interaction of the particles through the common radiation field leads to a shift of the natural frequencies of the system.

1. INTRODUCTION

WHEN considering the spontaneous emission by a system of particles which has dimensions smaller than the wavelength of the emitted radiation one has to take into account the coherence of the radiation emitted by the different particles.* The quantum mechanical account of this coherence in the spontaneous emission has been given in a number of papers.[2-5]

However, these papers did not take into account that each particle is located in the quasi-stationary induction zone of the field of all other particles. This will lead to a change in the state of the particles as compared to the state which they would be in in the absence of the interaction. Taking into account this interaction could lead for example to a change in the eigenfrequencies of the system. In references 3 and 5 only that part of the interaction was considered which leads to line broadening. A possible shift of the eigenfrequencies was not considered.

In view of the promises offered by apparatus which utilize the coherent spontaneous radiation in the radio-frequency range,[6,4,7,8] it seems advisable to investigate this question. The classical interaction energy of a system of charges is given up to terms of order v^2/c^2 by[9]

$$\mathcal{H}' = \sum_{A>B}\frac{e_Ae_B}{R_{AB}} - \sum_{A>B}\frac{e_Ae_B}{2c^2m_Am_BR_{AB}} \times[\mathbf{p}_A\cdot\mathbf{p}_B + (\mathbf{p}_A\cdot\mathbf{n})(\mathbf{p}_B\cdot\mathbf{n})]; \qquad (1)$$

here e_A is the charge, m_A the mass and $\mathbf{p}_A$ the momentum of a particle, R_{AB} the distance between particles, and $\mathbf{n} = \mathbf{R}_{AB}/R_{AB}$ the unit vector pointing from e_A to e_B. The first term of (1) gives the energy of the Coulomb interaction and will lead to the dipole interaction between the molecules. One can show that this term averages to zero for a large number of molecules. In the paramagnetic or ferromagnetic case this term vanishes for spherical specimens. One can easily see that the second term is important if the distances between the particles are shorter than the wavelength of the emitted radiation. If one considers the charge to be smeared out over the extent of the system then the second term of (1) can be approximately written as $(e^2/Mc^2R)\,\mathbf{P}^2$, where $\mathbf{P}$ is the momentum of the whole system, M its mass, R its radius, and e its charge. This term is of the same type as the kinetic energy of the system. One sees from this that connected with this term is a shift in the natural frequencies of the system. The next term in the expansion in powers of v/c (which has not been given here) will lead to radiative damping and has been treated in references 2 – 5.

In the present paper the interaction of the particles due to the common radiation field will be investigated.

2. CLASSICAL CONSIDERATIONS

We consider a system of particles, with spins and magnetic moments, located in a constant external magnetic field. In the following we shall speak, to be specific, of a system of electrons.

An electron in an external magnetic field H_0 has two energy levels associated with the two possible orientations of its magnetic moment — parallel and antiparallel to the magnetic field. We shall see later that the results obtained for the system of electrons in a magnetic field can almost completely be applied to an arbitrary system of quantum-mechanical objects with two energy levels.

For a sufficiently large number of electrons, one can assume that the magnetic moment of the system has a continuous distribution.

Now, according to Ginzburg[10] we can write the

*The necessity of considering this in classical theory was indicated in essence in the paper of Mandel'shtam.[1]

following system of equations determining the motion of the magnetic moment μ:

$$d\mu / dt = \gamma [\mu \times H_e] + \gamma \int [\mu \times H(x)] D(x) dv, \quad (2)$$

$$H = \text{curl}\, A; \quad \Box A = -4\pi\, \text{curl}\, M = 4\pi [\mu \times \text{grad}\, D],$$
$$M = \mu D(x); \quad \int D(x) dv = 1. \quad (3)$$

Here H_e is the external magnetic field, $H(x)$ the local magnetic field, $\gamma = -g\mu_B/\hbar$ the gyromagnetic ratio (μ_B is the Bohr magneton, $g = 2$ for a free electron), x the radius vector of a point in the electron system, M the magnetization vector, and $D(x)$ the distribution function of the magnetic moment. Elimination of the magnetic field leads to the following expression:[10]

$$\dot{\mu} = \gamma [\mu \times H_e] - (4\omega_m \gamma / 3\pi c^3) [\mu \times \ddot{\mu}] + (2\gamma / 3c^3) [\ddot{\mu} \times \dot{\mu}], \quad (4)$$

where $\omega_m = c/R$, R — a magnitude of the order of the radius of the system (or of another characteristic linear dimension).

We now consider the case where the external field H_e is sufficiently large. Then one can insert into the second and third terms of (4) for $\dot{\mu}$ the value following from $\dot{\mu} = \gamma \mu \times H_e$. If H_e is a constant field directed parallel to the z-axis then μ_x and μ_y are harmonic functions of time with a frequency $\omega_0 = |\gamma H_e|$ and $\mu_z = \text{const}$.

In the dipole approximation (when the dimensions of the system are much smaller than the wavelength of the emitted radiation) the radiation of the system will be fully determined by the variation of μ. It is easy to see that the second term leads to a shift of the emitted radiation with respect to ω_0 by the amount

$$\Delta_1\omega = (4\omega_m \omega_0^2 / 3\pi c^3) \gamma \mu_z, \quad (5)$$

while the third term produces a line broadening

$$\Delta_2\omega = \omega_0^3 \gamma \mu / 3c^3. \quad (6)$$

In deriving (6) it was assumed that μ_x, $\mu_y \ll \mu_z$ and $\mu_z \approx \mu$. The line broadening associated with the coherent spontaneous emission has been treated earlier in detail within the framework of quantum mechanics by the author.[4] Introducing the spin s measured in units of $\hbar$ corresponding to the magnetic moment $\mu = \gamma\hbar \mathbf{s}$, (4) describes the process of emission of quanta with energy $\hbar\omega_0$ associated with a varying quantum number m (projection of the spin on the z axis) and a constant spin s ($\sqrt{s(s+1)}$ is the absolute value of the magnitude of the spin vector).

For an arbitrary system of quantum-mechanical objects with two energy levels one can introduce the operator $\mathbf{R}$,[2,4,11] which is the analog of the total spin of the system. The emission will proceed without change of r (quantum-mechanical "cooperation number," the analogue of s) but by changing m, a quantity analogous to the projection of the spin on the z axis.

One can easily verify that Eq. (4) (without the second term) describes also the radiation process of any arbitrary system of quantum-mechanical objects which have two energy levels (even if an external magnetic field is absent). One just has to change γH_e to $\omega_0 = (E_2 - E_1)/\hbar$, $\mathbf{s}$ to $\mathbf{R}$ and $(g\mu_B)^2$ to $2|\mu_{12}|^2$ where μ_{12} is the matrix element of the (magnetic or electric) dipole moment of the particle. After this exchange we can compare (4) with the corresponding equation describing the change of m (see reference 4). Here it has been assumed that the condition $\omega_0 \gg \Delta_2\omega$ is fulfilled (the equations in reference 4 are given in the same approximation).

Thus the line width calculated by means of (4) has to coincide after the above indicated exchange with the line width of any arbitrary system of quantum-mechanical objects with two energy levels[4] (naturally for the case of large m and r where the classical and quantum-mechanical calculations have to coincide). Concerning the shift we shall show in the next section that the quantum-mechanical calculation also leads to $\Delta_1\omega$ of (5).

3. QUANTUM-MECHANICAL CALCULATION OF THE FREQUENCY SHIFT

The states of a system of weakly interacting quantum-mechanical objects with two energy levels can be described by two quantum numbers, r and m. Here $m = (n_+ - n_-)/2$ (with n_+ objects in the excited state and n_- in the ground state). If the interaction through the common field is not taken into account, the energy levels of the system are uniquely given by the quantum numbers m and ordered by the quantum number r. The energy of the system has a spectrum of the form $E_{rm} = m\hbar\omega$. We now calculate the level shifts due to the interaction of the system with the radiation field. The second-order term corresponding to emission and reabsorption of a photon of energy $k = \hbar\omega$ by the system gives a nonvanishing contribution:

$$W_{rm} = -\frac{2}{3\pi\hbar c^3} \int_0^K k dk \sum_{r'm'} |\dot{\mu}_{rm;\, r'm'}|^2 / (E_{r'm'} - E_{rm} + k). \quad (7)$$

Here $\hat{\mu}$ is the operator of the (electric or magnetic) dipole moment of the system, and $K = \hbar\omega_m$ the energy of the quantum with maximum possible frequency ω_m [this frequency coincides with the frequency $\omega_m = c/R$ introduced in (4)].

The matrix elements of the dipole moment of the system have the form[2,4]

$$|\mu_{rm;\,rm\mp 1}|^2 = |\mu_{12}|^2 (r \pm m)(r \mp m + 1), \qquad (8)$$

where μ_{12} is the matrix element of the transition $+ \rightarrow -$ for the particular quantum-mechanical object.

From (7) and (8) we find

$$W_{rm} = -\frac{2\omega_0^2}{3\pi\hbar c^3} |\mu_{12}|^2 \left\{ (r^2 - m^2 + r)\left(2K + \hbar\omega_0 \ln \left|\frac{K - \hbar\omega_0}{K + \hbar\omega_0}\right|\right) + m\hbar\omega_0 \left(\ln \frac{|K - \hbar\omega_0|(K + \hbar\omega_0)}{(\hbar\omega_0)^2}\right)\right\}. \qquad (9)$$

For $K \gg \hbar\omega_0$ (i.e., for a system with dimensions much smaller than the wavelength) we can write (9) in the form

$$W_{rm} = -(4\omega_0^2 / 3\pi\hbar c^3) |\mu_{12}|^2 (r^2 - m^2 + r) K. \qquad (10)$$

The energy of the level rm now equals $E_{rm} + W_{rm}$. The frequency shift corresponding the transition $m \rightarrow m-1$ is then given (for $m \gg 1$) by

$$\Delta_1\omega = (W_{rm} - W_{rm-1}) / \hbar = (4\omega_m \omega_0^2 / 3\pi c^3 \hbar) |\mu_{12}|^2 2m. \qquad (11)$$

This expression coincides with (5) which can be obtained in a classical way from (4) by replacing $(g\mu_B)^2$ with $2|\mu_{12}|^2$.*

4. CONCLUSION

The introduction of the quantum numbers r and m allows us to establish a certain correspondence principle, which states that to calculate the emission from a system† consisting of a sufficiently large number of quantum-mechanical objects having two energy levels, it is enough to calculate the classical equation, (4), describing the motion of a magnetic top in a magnetic field and then perform an appropriate substitution.

To illustrate this principle we juxtapose the classical [from Eq. (4)] and quantum mechanical formulae on the line shift and broadening‡ for a system of objects with two energy levels:

$$(\Delta_1\omega)_{cl} = \frac{4\omega_m\omega_0^2}{3\pi c^3} \gamma\mu_z, \qquad (\Delta_1\omega)_q = \frac{4\omega_m\omega_0^2}{3\pi c^3} \frac{2|\mu_{12}|^2}{\hbar} m,$$

$$(\Delta_2\omega)_{cl} = \frac{\omega_0^3\gamma}{3c^3} \mu, \qquad (\Delta_2\omega)_q = \frac{\omega_0^3}{3c^3} \frac{2|\mu_{12}|^2}{\hbar} r.$$

We see readily that the line shift (5) due to the interaction with the common radiation field is $\omega_m/\omega_0 \sim \lambda/R$ times greater than the corresponding line width of the radiation. This shift evidently can be experimentally observed in the radiation from ferromagnetic or paramagnetic materials or from a gas of molecules which have suitable levels in the radio-frequency range (where the condition $R \ll \lambda$ can be easily fulfilled).

We emphasize that we talk here about spontaneous radiation in free space. The radiation within resonators has an essentially different character (see, e.g., reference 11).

The radiation reaction and the interaction through the radiation field can achieve appreciable values in ferromagnetic materials (for example, ferrites),* which have a large concentration of electron spins.† Thus, for a ferrite with a volume of 10^{-3} cm^3 at $\lambda = 3$ cm we have $\Delta_2\omega \approx 10^8$ cps (the saturation magnetization is of the order 10^3 gauss).

Considering the radiation damping, we thus find that the relaxation time connected with the radiation is of the order 10^{-8} sec. It should be mentioned that for the case $\Delta_2\omega \ll \omega_0$ we can obtain from (4) the following equation‡ for the magnetization M [dropping the second term of (4)]:

$$d\mathbf{M}/dt = \gamma[\mathbf{M}\times\mathbf{H}] - (2\gamma^2\omega^2/3c^3) V [\mathbf{M}\times\mathbf{M}\times\mathbf{H}], \qquad (12)$$

where V is volume of the ferromagnetic sample. The dissipative term in this expression coincides with that of Landau and Lifshitz.[13]

In the considered example the frequency shift is of the order 10^9 cps. which is large enough to be observed experimentally.

However, the experiment has to be conducted in free space. The possibility of generation of radiation in free space is discussed in another paper by the author.[8]

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*This replacement is connected with the fact that in the classical case the oscillator is circularly polarized while in the present quantum-mechanical case it is linearly polarized. If the quantum-mechanical calculation is done for the case of circular polarization, we must replace $(g\mu_B)^2$ with $|\mu_{12}|^2$.

†We have here in mind a system of the type of an ideal gas (where the only interaction is through the radiation field).

‡The line width is calculated with the assumption μ_x, $\mu_y \ll \mu \approx \mu_z$. For the quantum-mechanical treatment the corresponding assumption is $r \approx |m|$. For the general case see ref. 4.

*An important characteristic of the ferrites is their small conductivity.

†A ferromagnetic material does not act like an ideal gas because of the strong exchange interaction. However, in a homogeneous ferromagnet the exchange forces do not have an effect on the equations of motion (4). This follows from the fact that the operator of the magnetic moment $\hat{\mu} = \sum_j \hat{\mu}_j$ commutes with the operator of the exchange energy $\sum_{ik} A_{ik}\hat{\mu}_i\hat{\mu}_k$ or, in the classical language, that the effective molecular field $H_M = qM$ is always parallel to the magnetization M. Therefore the torque $M \times qM$ vanishes (see ref. 12 on this).

‡In the case of ferromagnets we must consider H to be effective field.

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